

Anomalous Magnetic Properties of Sr_2YRuO_6

Ravi P. Singh and C. V. Tomy

Department of Physics, Indian Institute of Technology Bombay
Mumbai 400 076, INDIA

Abstract

Anomalous magnetic properties of the double perovskite ruthenate compound Sr_2YRuO_6 are reported here. Magnetization measurements as a function of temperature in low magnetic fields show clear evidence for two components of magnetic order ($T_{M1} \sim 32$ K and $T_{M2} \sim 27$ K) aligned opposite to each other with respect to the magnetic field direction even though only Ru^{5+} moments can order magnetically in this compound. The second component of the magnetic order at $T_{M2} \sim 27$ K results only in a magnetization reversal, and not in the negative magnetization when the magnetization is measured in the field cooled (FC) mode. Isothermal magnetization (M - H) measurements show hysteresis with maximum coercivity (H_c) and remnant magnetization (M_r) at $T \sim 27$ K, corroborating the presence of the two oppositely aligned magnetic moments, each with a ferromagnetic component. The two components of magnetic ordering are further confirmed by the double peak structure in the heat capacity measurements. These anomalous properties have significance to some of the earlier results obtained for the Cu-substituted superconducting $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ compounds.

1 INTRODUCTION

Sr_2YRuO_6 belongs to the family of double perovskite insulators, $\text{Sr}_2\text{LnRuO}_6$ (Ln = rare earth or Y) [1], where the Ru ions exist in the pentavalent state (Ru^{5+}) with a high-spin state $^4A_{2g}$ and $4d^3$ configuration ($J = 3/2$). Even though the structure of these compounds can be derived from the well known perovskite structure of SrRuO_3 by replacing alternate Ru ions with Ln ions [2], these compounds do not show any similarity to their parent compound SrRuO_3 , which is a ferromagnetic metal. The layered structure, consisting of alternate LnRuO_4 and SrO planes, accommodates both the Ru and rare earth atoms in the same LnRuO_4 plane and hence both the atoms share the same site symmetry (B -site of the perovskite structure ABO_3). The alternating positions of the Ru and Ln atoms in the unit cell result in two type of interactions between the Ru atoms; (i) direct interaction of Ru-O-O-Ru and (ii) indirect interaction through the rare earth atoms, Ru-O-Ln-O-Ru . Since the compounds having nonmagnetic Ln ions (Y and Lu) are also found to order magnetically [2, 3], the direct interaction is assumed to be stronger than the indirect interaction through the rare earth atoms. Among the $\text{Sr}_2\text{LnRuO}_6$ compounds, Sr_2YRuO_6 has captured additional interest due to the occurrence of superconductivity when Ru is partially ($\leq 15\%$) replaced by Cu [4, 8, 7, 5, 6, 9]. Cu is found to get substituted at the Ru site in the YRuO_4 planes and thus the structure of the substituted compounds remains the same as that of the parent compound, without creating any additional Cu-O planes [6].

The parent compound Sr_2YRuO_6 is known to be an antiferromagnetic insulator with the Ru moments ordering at $T_N = 26$ K [2]. The magnetic ordering temperature (T_N) was inferred as 26 K from the position of the peak in the magnetization measurements. Neutron diffraction measurements at 4.2 K have confirmed the magnetic ordering of the Ru moments, consisting of a type I AFM structure. Due to the monoclinic distortion of the structure, the compound is expected to show canting of the Ru moments resulting from the Dzyaloshinsky-Moriya (D-M) interactions [10, 11] among the antiferromagnetically ordered spins. How such a compound becomes a metallic magnetic superconductor without creating Cu-O planes is still a puzzling question. There are still many unanswered questions regarding the origin of magnetism and superconductivity in the Cu-substituted Sr_2YRuO_6 compounds. At the same time, there are no detailed magnetization studies available for the parent compound itself, except for one report on Sr_2YRuO_6 single crystals [12] which confirms the magnetic ordering and weak ferromagnetism. In addition, the resistivity of Sr_2YRuO_6 single crystals [12] shows anomalous behaviour below T_N followed by a Mott-like transition at 17 K whereas the magnetoresistance becomes negative below 30 K. The band structure calculations [13] have indicated the competition between the antiferromagnetic and ferromagnetic fluctuations among the Ru moments. We present here some additional evidence for the competition be-

tween antiferromagnetic and ferromagnetic coupling in this compound. Detailed measurements of magnetization and heat capacity show some anomalous properties exhibited by Sr_2YRuO_6 . Both the measurements unfold clear evidence for two magnetic orderings ($T_{M1} \sim 32$ K and $T_{M2} \sim 27$ K), even though the magnetic ordering in this compound can occur only by Ru moments. The magnetization measurements corroborate that both the magnetic ordering occurs with ferromagnetic components and these two components align opposite to each other with respect to the magnetic field direction, resulting in a magnetization reversal. The results presented here have relevance to the magnetic properties exhibited by the Cu-substituted superconducting $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ compounds [4, 8, 7, 5, 6, 9].

2 EXPERIMENTAL DETAILS

Polycrystalline samples of Sr_2YRuO_6 were prepared by the standard solid state reaction method by mixing stoichiometric amounts of SrCO_3 , Y_2O_3 and Ru metal powder and heating at 960°C for 12 hours. The final sintering of the pelletized powder was carried out at 1360°C for 24 hours after several intermediate heat treatments followed by grindings. X-ray diffraction pattern of the samples was recorded on an X’pert PRO diffractometer (PANalytical, Holland). The magnetization as a function of temperature and magnetic field was measured using a vibrating sample magnetometer (Quantum design, USA). The heat capacity measurements using the relaxation method were performed using a physical property measurement system (Quantum design, USA) in the temperature range 1.8-300 K.

3 RESULTS AND DISCUSSION

The Rietveld analyses of the x-ray diffraction patterns using Fullprof software showed that the compound forms in single phase with a monoclinic structure (space group $P2_1/n$). The lattice parameters obtained from the analyses are, $a = 5.769$ Å, $b = 5.772$ Å and $c = 8.159$ Å along with $\beta = 90.18^\circ$ which are in good agreement with those reported earlier [2]. Figure 1 illustrates the magnetization of Sr_2YRuO_6 as a function of temperature in zero field-cooled (ZFC) and field-cooled (FC) modes. In the ZFC measurements the sample was cooled in zero applied field to 2 K, the required magnetic field was applied and then the data were taken while increasing the temperature. For the FC measurements, the sample was cooled from the paramagnetic state to 2 K in an applied field and the data were recorded while heating the sample. In order to minimize the remnant field in the superconducting magnet before the ZFC measurements, the magnetic field was reduced to zero from a large field value in the oscillating mode. This made sure that the remnant field was within

± 2 Oe. The lower panel shows the ZFC measurements for various applied field values. For low field values, the magnetization is negative at lower temperatures. As the temperature is increased, the magnetization remains independent of temperature till ~ 20 K and then surprisingly decreases to go through a minimum. As the temperature is further increased, the magnetization increases, goes through a positive maximum and then shows the normal paramagnetic behaviour. For $H = 1.5$ kOe, the ZFC magnetization starts with a positive value at low temperatures, but goes through negative value at the minimum. For higher fields, the magnetization is always positive, even though it goes through a minimum. The width of the peaks at the maximum and minimum as well as the temperature at which they occur depends slightly on the applied fields; both decrease with increasing field. The upper panel of Fig. 1 shows the FC measurements at various applied fields. The FC magnetizations show a broad peak and the temperature at which the peak occurs shows a weak temperature dependence on the applied fields.

In order to ascertain that the anomalies observed in the ZFC magnetization is not entirely due to effects of negative remnant magnetic field in the superconducting coils, we have carried out FC measurements in smaller field values, both positive and negative. Figure 2 shows the FC measurements for an applied field of ± 10 Oe. It is clear that the FC magnetization remains negative whether the field is positive or negative. Such effects are seen upto 25 Oe above which the FC curves switch over to the positive side. Neutron diffraction studies at 4.2 K [2] had indicated only an AFM ordering of the Ru moments. It was also proposed that the distorted monoclinic structure can give rise to a small canting of the Ru moments and hence a small ferromagnetic component in this compound resulting from the D-M interactions between the antiferromagnetically ordered Ru moments. This, however, cannot explain the observed magnetization behaviour in this compound. A simple ferromagnetic component due to canting can make the magnetization negative in the ZFC mode if the remnant field is negative. But then the magnetization will monotonically decrease as the temperature is increased and will cross over to the positive side before completing the magnetic order. A typical example for such a behaviour is shown in the inset of Fig. 2 for MnCO_3 , which is a well known canted antiferromagnet having D-M interactions [14]. We have also made sure that the compound does not contain any SrRuO_3 impurities (not detected in x-ray diffraction patterns) by taking the ZFC and FC data for small field values in the temperature range 100-200 K (even a small trace of SrRuO_3 impurity will give a thermal hysteresis around its ferromagnetic ordering temperature (150-160 K) between the ZFC and FC measurements).

In order to further corroborate that there exists more than a simple canting in this compound, we have carried out detailed magnetization measurements as a function of magnetic field at different temperatures. Figure 3 depicts the low field magnetic isotherms at some selected temperatures. At low temperatures, well

below the magnetic anomalies, the magnetization curves are almost linear with a small value for coercivity (H_c). As the temperature crosses 22 K, the magnetization shows significant hysteresis and the magnetization loops open up. The opening of the loop increases until the temperature reaches ~ 27 K and then decreases as the temperature is further increased. Even at 32 K, the hysteresis is much more than the same at 5 K. At 35 K, we see only a linear behaviour expected for a paramagnet. Even though the hysteresis loops are not closed at some temperatures (Fig. 3(d), 3(e)), they show a normal behaviour when the applied fields are extended to higher values (see main panel of Fig. 4). No other anomalies are observed in the high field magnetization curves. The coercivity and the remnant magnetization plotted as a function of temperature in the inset of Fig. 4 show a maximum near 27 K and decrease on either side of this temperature. This clearly demonstrates that some sort of magnetization reversal happens at 27 K.

There are no reports about the heat capacity of this compound in the literature. The result of our heat capacity measurements for Sr_2YRuO_6 is presented in Fig. 5(a). Two peaks are obvious, one at $T \sim 30$ K and the other at $T \sim 26$ K, which correspond well to the anomalies observed in the magnetization. There is only a minor effect by the magnetic field on the heat capacity of the sample even at 50 kOe (Fig. 5(a)), even though a small decrease in the temperature dependence of the peak positions were observed in the magnetization measurements. In order to have an estimate of the approximate magnetic heat capacity, the phonon contribution needs to be subtracted from the total measured heat capacity. Since there are no nonmagnetic analogues available for this compound, the phonon contribution was calculated from the combined Debye and Einstein equations [15],

$$C_{ph} = R \left(\frac{1}{1 - \alpha_D} \left(\frac{\theta_D}{T} \right)^3 \int_0^x \frac{x^4 e^x}{(e^x - 1)^2} dx + \sum_{i=1}^{3n-n} \frac{1}{1 - \alpha_E} \frac{(\theta_{Ei}/T)^2 \exp(\theta_{Ei}/T)}{(\exp(\theta_{Ei}/T) - 1)^2} \right) \quad (1)$$

where α s are the anharmonicity coefficients, θ_D is the Debye temperature, θ_E is the Einstein temperature and $x = \theta_D/T$. The best possible fit was obtained when the calculations were performed by using one Debye and three Einstein frequencies along with a single α_E . The solid line in Fig. 5(b) represents the fit to the phonon contribution, which is in good agreement with the experimental data at high temperatures (above the magnetic ordering). The parameters obtained from the best fit are: $\theta_D = 200$ K, $\theta_{E1} = 300$ K, $\theta_{E2} = 529$ K, $\theta_{E3} = 615$ K, $\alpha_E = 1.0 \times 10^{-4}$ K and $\alpha_D = 1.0 \times 10^{-4}$ K. The Debye and Einstein temperatures obtained for Sr_2YRuO_6 are comparable with those obtained for YVO_3 where the phonon contribution was obtained in a similar fashion, but with only two Einstein frequencies along with one Debye frequency [16]. The magnetic heat capacity, C_{mag} , obtained by subtracting the calculated phonon heat capacity from the total heat capacity, is shown in

the Fig. 5(c) along with the magnetic entropy ($S_{mag} = \int_{T_1}^{T_2} \frac{C_{mag}}{T} dT$). The two peaks become more obvious in the magnetic heat capacity. In Sr_2YRuO_6 , the magnetic transition can occur only due to the ordering of Ru^{5+} moments. In that case, the exact reason for the observed double peak behaviour is not very clear at present. Magnetic entropy (S_{mag}) increases with temperature and saturates to a value of $\sim 2.6 \text{ J mole}^{-1} \text{ K}^{-1}$ above 30 K. If we consider the ground state of Ru^{5+} ions as $J = 3/2$, then the expected magnetic entropy is $11.52 \text{ J mole}^{-1} \text{ K}^{-1}$ ($S = R \ln[2J + 1]$), corresponding to the four-fold degenerate ground state. However, the crystalline electric fields, if present, can split this ground state into two doubly degenerate states, giving rise to a ground state multiplicity of only two [17]. This will reduce the magnetic entropy of the compound to $5.76 \text{ J mole}^{-1} \text{ K}^{-1}$ ($S = R \ln 2$). The observed entropy, however, is even less than half of this value. In fact, neutron diffraction measurements had estimated a value of $1.8 \mu_B/\text{Ru}^{5+}$ at 4 K (instead of the expected value of $3 \mu_B/\text{Ru}^{5+}$) in the magnetically ordered state [2]. This moment value corresponds well with the doubly degenerate ground state. If we compare the reduction in entropy of Sr_2YRuO_6 to that observed for YVO_3 [16], frustrations of Ru spins at high temperatures (above the magnetic ordering) can be attributed as the reason for the reduction in entropy. The correlation between the frustrated moments at high temperatures reduces the contribution of the entropy to the magnetic ordering. Such a frustration among the Ru moments is inferred in Sr_2YRuO_6 as the possible reason for the reduction in T_N even though the compound possesses a large exchange integral value [13].

It is clear that Sr_2YRuO_6 exhibits two anomalies, the first at $\sim 32 \text{ K}$ (T_{M1}) and the second at $\sim 27 \text{ K}$ (T_{M2}) even though the magnetic ordering in this compound can come only from the Ru^{5+} moments. If we assume that the two anomalies are associated with the magnetic ordering of the Ru moments, then the observed behaviour is very interesting. The isothermal magnetization curves at different temperatures (Fig. 3) clearly demonstrate that the first magnetic order starts at $T_{M1} \sim 32 \text{ K}$ with a ferromagnetic component resulting in the increase of hysteresis and H_c as the temperature is lowered. This ferromagnetic component is expected from the canting of the antiferromagnetically ordered Ru moments because of the D-M interactions. However, the decrease of hysteresis and H_c below 27 K indicates that a second component of the magnetic order also develops with a ferromagnetic component (T_{M2}), but aligns itself opposite to the first component and hence opposite to the applied field. This component almost cancels the first component and hence the hysteresis is negligible at low temperatures ($< 20 \text{ K}$). These anomalies are further confirmed in the zero field remnant magnetization measurements as shown in Fig. 6(a). Here the sample was cooled (FC) in a field of 5 kOe down to 10 K. The field was then removed and the remnant magnetization was measured in zero field while warming the sample. The remnant magnetization shows a normal decrease upto $\sim 20 \text{ K}$, but then increases, goes through a maximum at $\sim 27 \text{ K}$ and then decreases to zero

above 32 K. This clearly demonstrates that the magnetic ordering consists of two components and they are aligned opposite to each other with respect to the magnetic field direction. While cooling the sample in magnetic field, the first component orders and aligns parallel to the field at ~ 32 K, but the second component aligns antiparallel to the field at ~ 27 K, decreasing the net magnetization. However, this antiparallel component is not strong enough to make the magnetization negative as in the case of some $LnVO_3$ compounds. As the sample is warmed up in zero field, the remnant magnetization increases when the antiparallel component relaxes and completes its disordering.

In order to verify the thermodynamic reversibility of the FC state, we have measured the remnant magnetization by field-cooling the sample down to two pre-selected temperatures (T_1 and T_2) on either side of the maximum in the FC magnetization curve as shown in Fig. 6(a). In the first case, the sample was field cooled to 28 K ($T_{M2} < T_1 < T_{M1}$), where only the first magnetic component would have ordered, and the field was removed. In the second case, the sample was field cooled to 23.5 K ($T_2 < T_{M1}, T_{M2}$), where both components would have undergone magnetic ordering, before removing the field. In both cases, the remnant magnetization follows exactly the original remnant curve which was obtained by switching the field off at 10 K, proving the thermodynamic reversibility of the magnetically ordered states. Further evidence for the opposite alignments of the two components of magnetic ordering is evident from the data in Fig. 6(b). Here the sample was cooled in a negative field (-50 Oe) so that the magnetization at 5 K is negative. At 5 K, the field was increased in the positive direction (~ 2.2 kOe) until the magnetization became positive. Magnetization shows normal behaviour upto 20 K, but shows a sudden dip to go through a negative minimum at ~ 27 K. Thus it is clear that whether the sample is cooled in positive or negative field, the two components of magnetic order align always opposite to each other.

Neutron diffraction measurements in this compound at 4.2 K [2] had indicated that the nuclear structure of the compound remains unchanged at 4.2 K, ruling out the possibility of any structural changes. On the other hand if we assume that one of the anomalies is due to a possible structural change, then the changed structure should again revert back to the original structure at low temperatures. From the position of the magnetic diffraction peaks, magnetic ordering of the Ru moments was deduced to be antiferromagnetic with a type I structure [2]. However, the temperature variation of the intensity of magnetic peak(s) was not reported and hence the exact temperature at which the Ru moments order is not available. The T_N of ~ 26 K was assigned to this compound simply from the position of the peak in the high field magnetization measurements [2]. Even though there are no detailed experimental observations in Sr_2YRuO_6 , many experimental results including neutron diffraction exist for Cu-substituted $Sr_2YRu_{1-x}Cu_xO_6$ compounds [7, 6, 9, 8]. Assuming that the magnetic properties associated with the ordering of the Ru moments

are not drastically altered, we can analyze the results of the μ SR measurements in the Cu-substituted compounds [9, 7]. Both the precession frequency and relaxation rate show anomalies at ~ 30 K for muons trapped in the two possible sites; oxygen in the YRuO₄ layers ($\mu_{O(1,2)}$) and oxygen in the SrO layers ($\mu_{O(3)}$). The authors assigned this anomaly to the fluctuation of the Ru moments which order in a spin-glass state. This is exactly the same temperature range at which the first component of magnetic ordering starts in our studies for the parent compound (T_{M1}). The variation of the power exponent at the $\mu_{O(1,2)}$ site as a function of temperature [9] almost resembles the mirror image of our temperature variation of the coercivity and remnant magnetization (inset of Fig. 4). It was also proposed that the fluctuations of the Ru moments stop at $T = 23$ K and order antiferromagnetically. We have identified this temperature in our studies as the temperature at which the second component of magnetic moments completes its ordering. ^{99}Ru Mössbauer measurements in $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ ($x = 0.05$) [7, 8] have confirmed the magnetic ordering of Ru moments. Both the isomer shift and the hyperfine field values are consistent with the pentavalent nature of the Ru moments. From the temperature variation of the Mössbauer spectra [8], it was concluded that the magnetic ordering of the Ru moments persists upto 30 K, which is consistent with the magnetic ordering temperature T_{M1} in our measurements. Additional support for the Ru ordering as high as 30 K comes from the neutron diffraction measurements reported for $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ ($x = 0.15$) where the intensity of the magnetic peak due to the ordering is Ru moments is evident upto ~ 30 K [6].

In ZFC measurements, the sample is always cooled in a nominal remnant field ($\sim \pm 2$ Oe in our case). This may result in the negative magnetization at low temperatures. When the magnetic field is applied, the magnetization remains negative for low fields (Fig. 1) but changes over to positive values for sufficiently large fields. Even then a magnetization reversal happens and the magnetization goes through a negative minimum in between the two transitions. This behaviour may be caused by the fact that the large fields flip (to orient parallel to the field) some, but not all, of the spins oriented against the field. As the field value increases, majority of the spins, oriented antiparallel to the field, align parallel to the field, resulting only in a small minimum at T_{M2} . In FC magnetization ($H \geq 50$ Oe), the first component of the magnetic transition aligns along the field at T_{M1} , resulting in a positive magnetization. However, at T_{M2} , the second component aligns antiparallel to the first component, resulting in the reduction in magnetization. When this alignment is over, the magnetization remains constant, as in the case of FC measurements in smaller fields.

The reason for the two components of the magnetic ordering reminiscent of ferromagnetic ordering in this compound is not clear. In the ordered double perovskite $\text{Sr}_2\text{LnRuO}_6$ compounds, the B-site of the perovskite structure ABO_3 is uniquely occupied either by Ru or the rare earth metal ions due to the lower coordination

number compared to Sr. Since Ruthenium is considered to be in the oxidation state of 5+ in these compounds, the chances of ferrimagnetic ordering of $\text{Ru}^{5+}/\text{Ru}^{4+}$ moments are very rare. We have repeated the measurements with samples annealed in air, oxygen or argon and all of them showed the same behaviour. In fact, the isomer shift values from the ^{99}Ru Mössbauer measurements in Sr_2YRuO_6 had confirmed the pentavalent oxidation state of Ru moments [7, 18]. Even if the spin-glass state is assumed as suggested by Harshman et al [9], the observed properties - the magnetization reversal, thermodynamic reversibility of magnetization and two peaks in heat capacity - cannot be explained. The magnetic interactions among the ordered Ru moments can take place in two ways; (i) the σ -super exchange interaction between nearest neighbour (nn) Ru^{5+} ions via Ru-O-O-Ru pathway and (ii) the π -super exchange between the next nearest neighbour (nnn) Ru^{5+} ions via Ru-O-Y-O-Ru pathway. The relative strengths of these two interactions will determine the type of magnetic ordering at low temperatures. Since Y is a nonmagnetic ion, it is not expected to take part in the exchange interaction and hence the second interaction between the nnn is assumed to be negligible. This assumption can further be supported by the fact that the relaxation rate at the $\mu_{\text{O}(3)}$ site for the muons is very much smaller than the relaxation rate at the $\mu_{\text{O}(1,2)}$ site [9, 7] since the Ru-O-Y-O-Ru pathway includes the oxygen at the O(3) sites only. Whether the competition between these two interactions, however, can give rise to the observed double peak behaviour needs further investigations. It is possible that the stronger interaction orders the Ru moments at T_{M1} and the weaker interaction realigns some of the Ru moments in the opposite direction at T_{M2} . It is possible that the second component of the magnetic order at T_{M2} is not strong enough to bring the magnetization down to negative values even though it gets reduced. However, why the second component aligns the moments against the first component and hence the magnetic field is not clear now. Detailed neutron diffraction measurements are needed as a function of temperature to explain the reason for the observed anomalies.

In conclusion, we have reported some of the anomalous properties exhibited by SrYRuO_6 deduced from the detailed magnetic and heat capacity measurements. Two distinct magnetic orderings are evident in the compound even though only Ru moments can order in this compound. The two components of the magnetic order align always opposite to each other and to the magnetic field. Observation of hysteresis and coercivity in magnetic isotherms below the magnetic order indicates the presence of ferromagnetic component associated with the magnetic ordering. The presence of two well defined peaks in the heat capacity indicates that the two magnetic components have large entropy change associated with the ordering.

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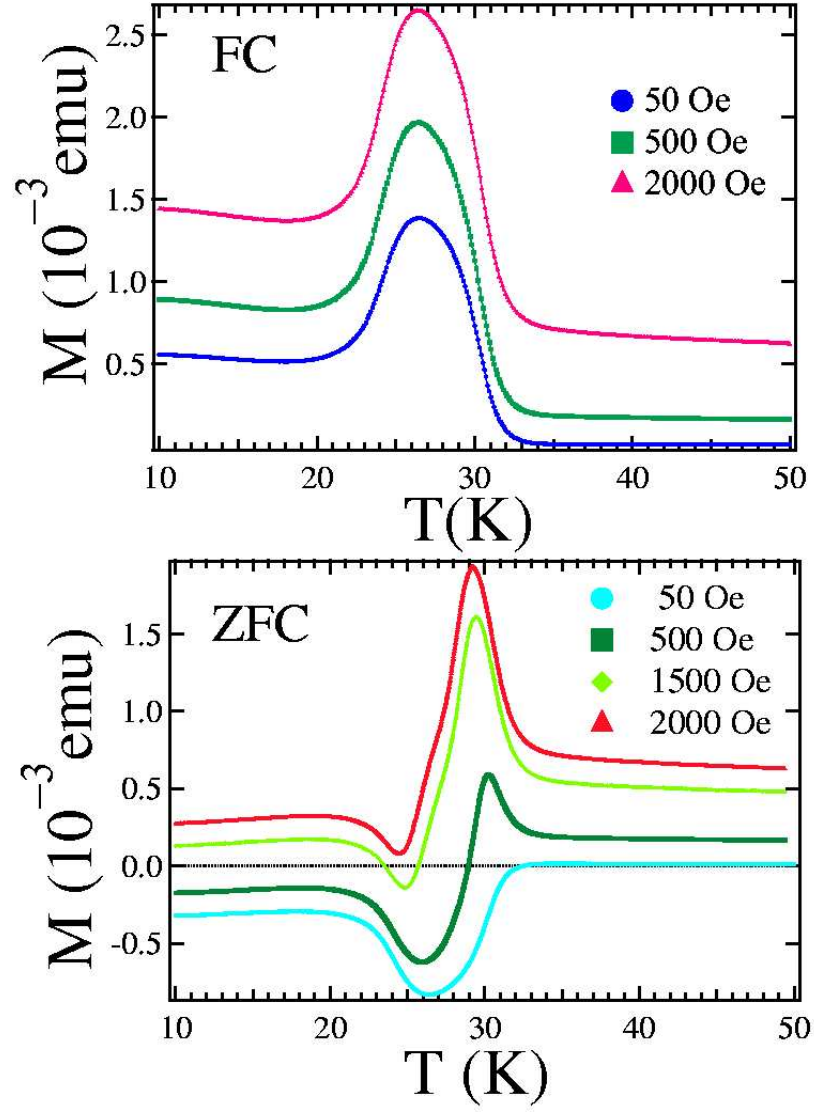


Figure 1: Magnetization vs temperature for Sr_2YRuO_6 in zero field-cooled (ZFC) and field-cooled (FC) modes under various applied fields.

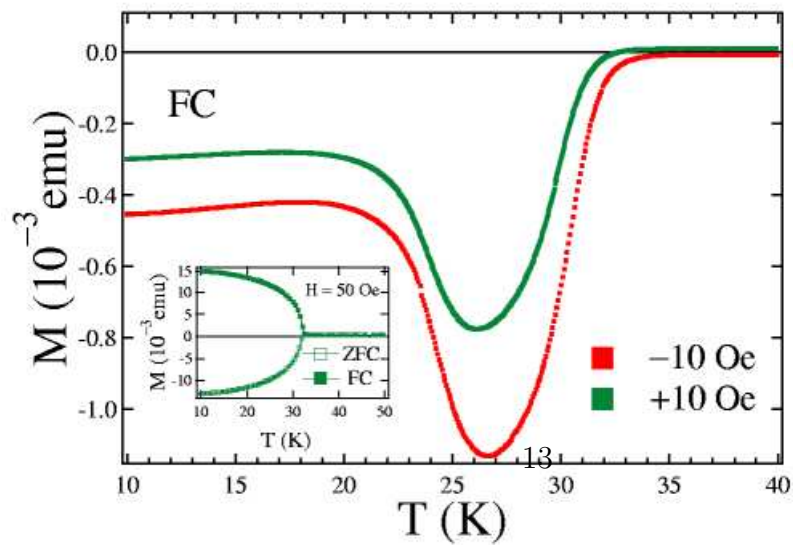


Figure 2: Magnetization for Sr_2YRuO_6 in FC mode at ± 10 Oe. Inset shows the FC and ZFC curves for MnCO_3 .

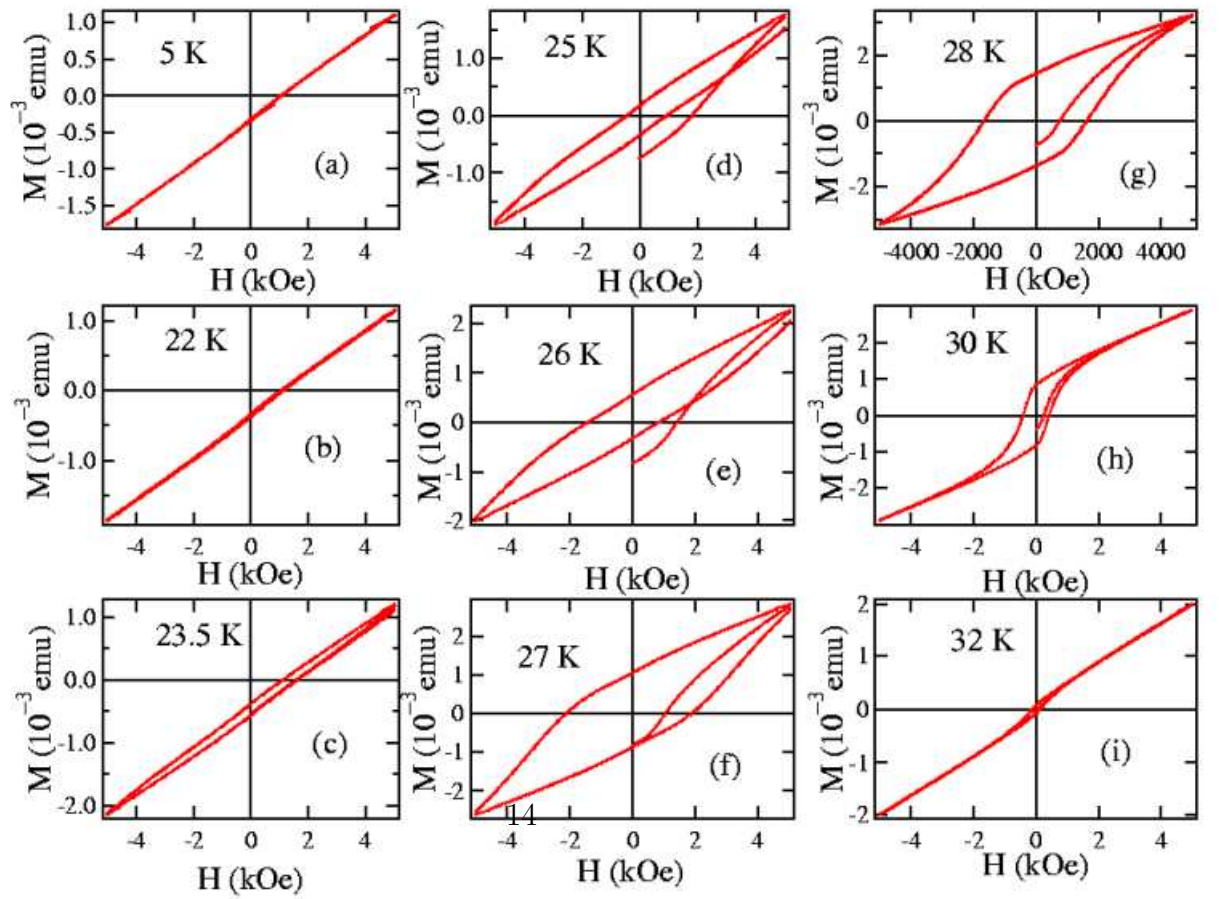


Figure 3: Isothermal magnetization curves for Sr_2YRuO_6 at different temperatures.

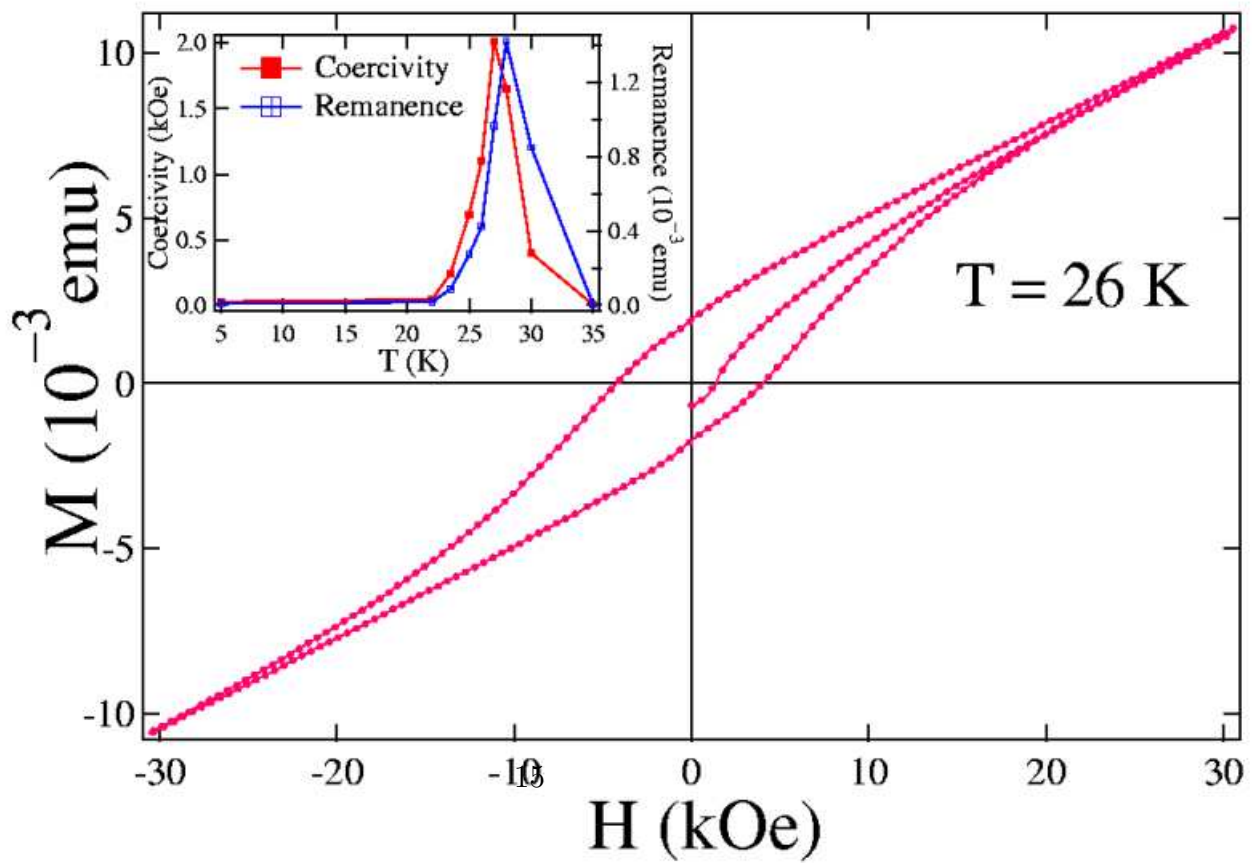


Figure 4: High field magnetization as function of field at 26 K. Inset shows the temperature variation of coercivity (H_c) and remanence (M_r)

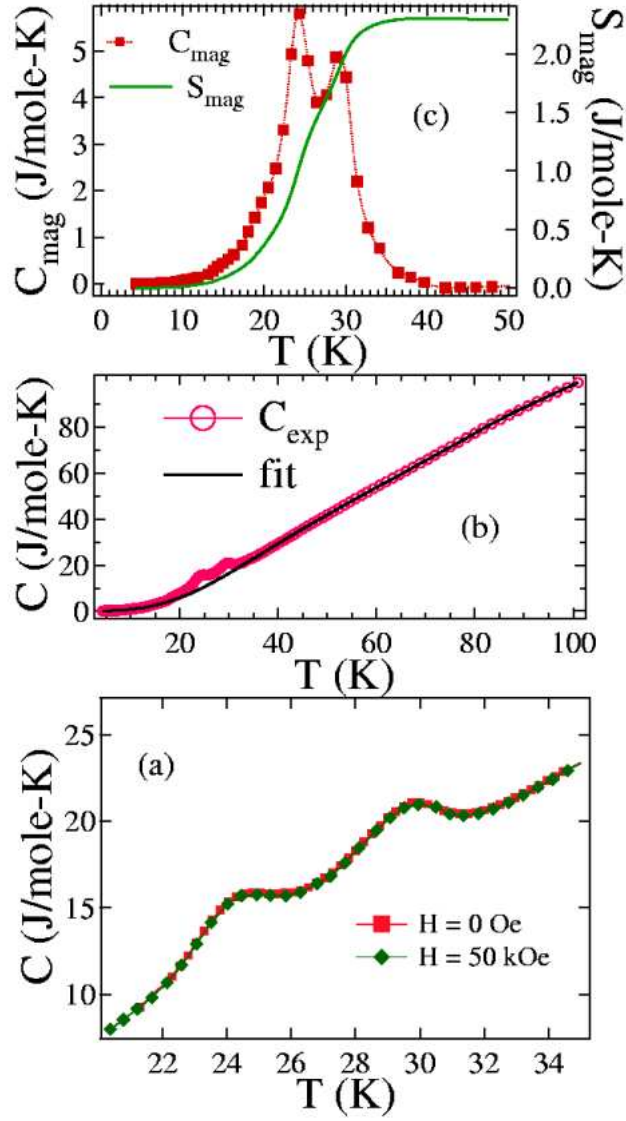


Figure 5: (a) Measured total heat capacity (C_v) of Sr_2YRuO_6 in applied fields $H = 0$ Oe and 5 kOe. (b) Heat capacity with the fit (solid line) for phonon contribution. (c) Magnetic contribution to heat capacity (C_{mag}) (left scale) and calculated magnetic entropy (S_{mag}) (right scale) as a function of temperature.

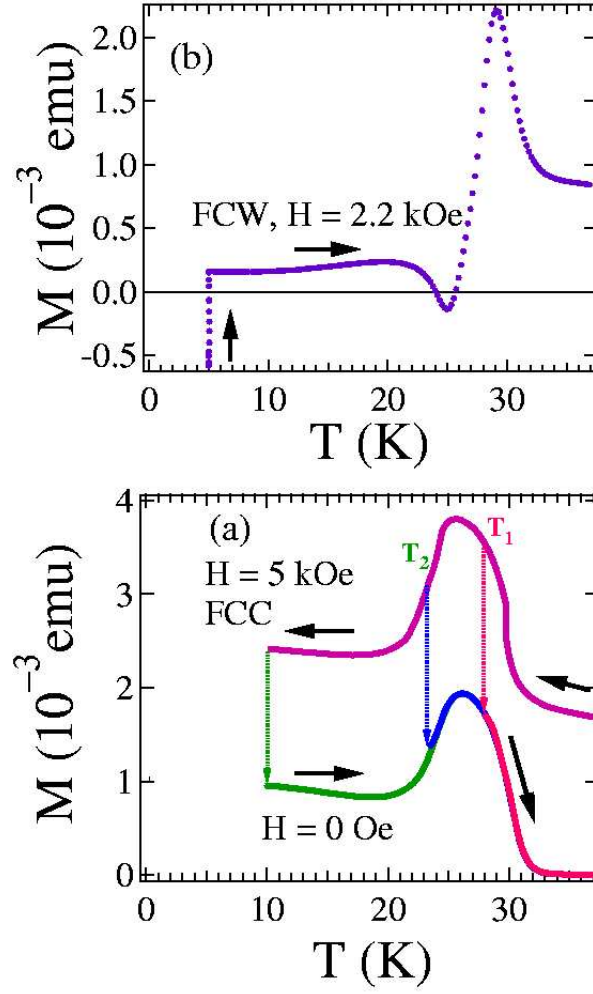


Figure 6: (a) FC data while cooling the sample in an applied field of 5 kOe (top curve) and the zero field warm up data (bottom curves) after switching off the applied magnetic field at $T = 10$ K, 23.5 K and 28 K. (b) FC warm up data in an applied field of 2.2 kOe after cooling the sample in a negative applied field of -50 Oe.